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CONTINUOUS AIR MONITOR FOR ALPHA-EMITTING AEROSOL PARTICLES

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Abstract

A new alpha CAM sampler is being developed for use in detecting the presence of alpha-emitting aerosol particles. The effort involves design, fabrication and evaluation of systems for the collection of aerosol and for the processing of data to speciate and quantify the alpha emitters of interest. At the present time we have a prototype of the aerosol sampling system and we have performed wind tunnel tests to characterize the performance of the device for different particle sizes, wind speeds, flow rates and internal design parameters. The results presented herein deal with the aerosol sampling aspects of the new CAM sampler. Work on the data processing, display and alarm functions is being done in parallel with the particle sampling work and will be reported separately at a later date.

Wind tunnel tests show that ≥50% of 10 μm aerodynamic equivalent diameter (AED) particles penetrate the flow system from the ambient air to the collection filter when the flow rate is 57 L/min (2 cfm) and the wind speed is 1 m/s. The coefficient of variation of deposits of 10 μm AED aerosol particles on the collection filter is 7%. An inlet fractionator for removing high mobility background aerosol particles has been designed and successfully tested. The results show that it is possible to strip 95% of freshly formed radon daughters and 33% of partially aged radon daughters from the aerosol sample. This offers the possibility of improving the signal-to-noise ratio in the alpha energy spectrum region of interest thereby enhancing the performance of background compensation algorithms.

I. Background

Alpha continuous air monitors (CAMs) are used in the nuclear industry to detect the presence of transuranic (TRU) alpha-emitting aerosol particles. In principal, a steady flow of air is drawn into the CAM sampler and the aerosol particles are deposited on a collection substrate where the radioactivity energy spectrum is continuously monitored. Generally the particles are separated from air by filtration, although inertial impaction has also been used for collection (Tait, 1956; Alexander, 1966). In the case of a filter collector, the detector is placed parallel to the filter at a distance of approximately 5 mm from the filter face. Typically, with an inertial impactor sample deposition takes place on a substrate located over the detector.

Ideally, each radioisotope has a unique alpha energy signature which should render the speciation and quantification process straightforward. However, there are several practical limitations which manifest themselves in obscuring the true results. First, in the case of filter detectors, the air gap causes a distortion of the low-energy tails of the alpha peaks leading to severe overlap, which suggests that CAM samplers should incorporate designs which reduce the gap to as mall a value as practical. There is a limitation which must be taken into account since, if the gap is reduced below a certain level, there will be inadvertent losses of aerosol particles on the internal sampler walls in the filter/detector region. In a unique device for dealing with this problem, Kaifer et al. (1986) separated the sampling and readout functions of a CAM and collected the aerosol at ambient pressure and performed the analysis under vacuum in order to improve the resolution of the energy spectra. Due to the additional complexity and high cost of implementation, a vacuum readout approach was not considered in the present design.

Second, in the case of inertial impactors, the mechanics of operation preclude the collection of particles with sizes $\leq 0.5\ \mu m$ which, for certain types of aerosol release mechanisms, can cause a failure to detect over half of the alpha-emitting aerosol particles present since, in some sampling situations, the mass median aerosol size is $<0.5\ \mu m$ AED (Kirchner, 1966; Elder et al., 1974). Also, inertial impactors have an inherent tendency to cause large particles to rebound from the collection surface and be carried away with the exhaust air stream. Greasing the collection surface will reduce the problem, however, the grease layer will cause additional distortion of the energy spectrum.

Third, the presence of alpha emitting background radionuclides (radon and thoron progeny) can cause difficulty in recognition of TRUs at concentrations far above regulatory alarm levels. For example, Pu-239 emits alpha radiation with an energy of 5.15 MeV and RaA/ThC emits at 6.0 MeV. Thus, the Pu region of interest in the alpha energy spectrum lies in the low energy tails of the natural background peaks. At the alarm level concentration for Pu-239 given in U.S. DOE Order 5480.11, the typical CAM sampler will register about 15 cpm from the TRU and may detect an order or two of magnitude greater count rate in the same region of interest after 8 hours of sampling due to the tailing of the energy spectra of the radon/thoron progeny. The most common approach to dealing with this problem is to employ a numerical algorithm to subtract an estimation of the background counts from the TRU energy channels. But, background compensation has definite limits in high background conditions. An alternative approach is to try to eliminate some of the background radionuclides from the sample. If the radon/thoron progeny are relatively free from attachment to other aerosol particles, some separation of the background radioactivity can be accomplished prior to collection of the aerosol. For example, in a fractionating CAM sampler head design based on inertial impaction alone, the mobile background aerosol particles are separated and carried away from the collection substrate along with the fine (usually submicrometer) fraction of the aerosol. For a filter collector design, the fractionation must be performed upstream of the filter, since an aerosol sampling filter will have a efficiency that approaches 100% for all particle sizes including those of freshly-formed radon/thoron progeny. In the development of our CAM sampler, we have incorporated fractionation stages upstream of the filter to strip freshly-formed, highly mobile radon/thoron progeny from the size distribution. Preliminary experiments have been conducted to demonstrate the feasibility of the concept.

A fourth problem of many contemporary CAM samplers is that an unbiased sample of aerosol does not reach the collection substrate. Losses on the internal walls of the sampler can substantially reduce the concentration of large particles detected by the CAM. In a previous study, we conducted wind tunnel tests with CAM samplers supplied by three vendors in which we examined the penetration of 5, 10, and 15 μm AED aerosol particles from the free stream to the sampling filters (McFarland et al., 1990). For one of the units, essentially no particles with sizes larger than 6 μm AED were able to penetrate through the flow system to the filter. One goal of the present development is that the CAM should permit penetration of at least 50% of aerosol particles of 10 μm AED. This size was selected by the U.S. EPA (1987) as representing the division between aerosol which could penetrate to the thoracic region of the human lung (≤ 10 μm AED) and that which would be deposited in the extra-thoracic regions.

A fifth factor, which will cause problems in determining the concentration of RDUs is that of non-uniformity of filter deposits. If aerosol particles are predominantly deposited near the edge of a filter, the counting efficiency will be reduced and the CAM will underestimate the concentration (Rodgers and McFarland, 1989). Should the deposit be primarily in the center of the filter, there would be an overestimation of concentration. Biermann and Valen (1983) tested a commercially available CAM sampler and observed substantial non-uniformities. In the study of three commercially available CAM samplers, McFarland et al. (1990) examined the areal deposition of 10 μm AED particles by analyzing subsamples cut from sampling filters. The coefficients of variation of areal deposition were 18% and 39% for the two units which did transmit significant concentrations of 10 μm AED aerosol particles.

In the present CAM sampler development, we have designed and fabricated a prototype which has been wind tunnel tested to determine the aerosol transport characteristics. Tests have been conducted to determine filter uniformity. Also, bench-type studies have been performed with freshly-formed radon daughters to ascertain the feasibility of using fractionators to separate the highly mobile background fraction from the distribution.

II. Prototype CAM

The prototype CAM sampler is shown schematically in Figure 1. Wind tunnel testing of the unit has been performed without the electronics assemblies, since the design and placement of those components does not interact with the aerosol flow path in the CAM. With reference to Figure 1, aerosol at a design flow rate of 57 L/min is drawn into the CAM though a diffusion screen system which is intended to serve two functions; namely, to collect highly mobile unattached background alpha-emitters and to uniformize the aerosol velocity profile. Furtner inside the unit, the air is directed through an electrical field in a condenser. The condenser is intended to provide essentially quantitative collection of charged

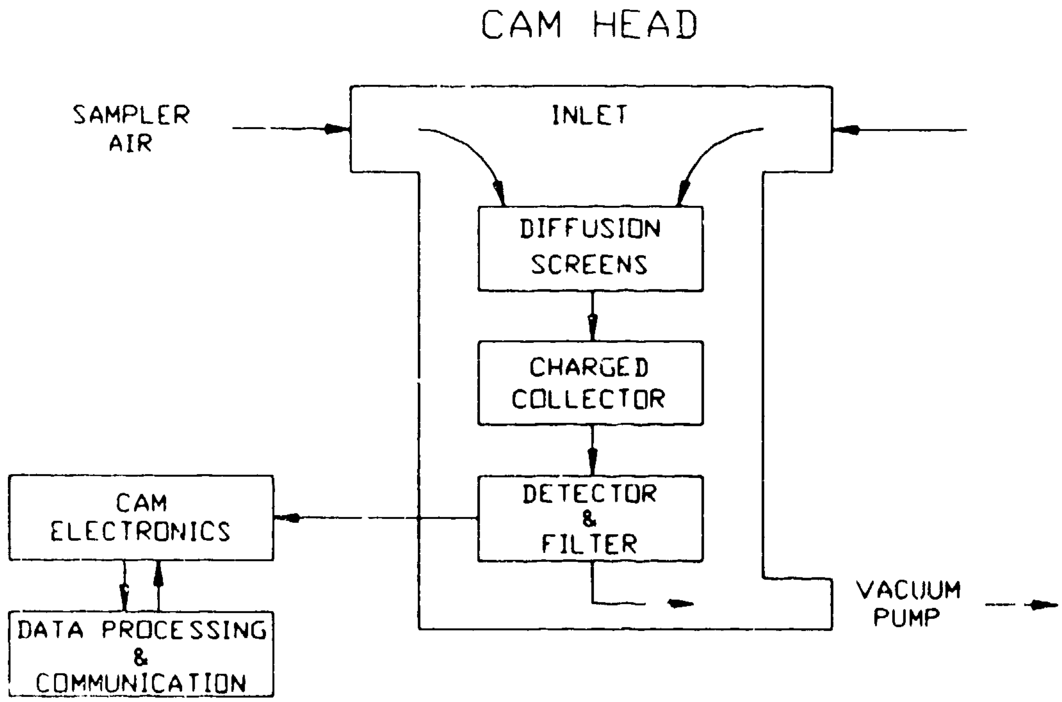


Figure 1. Schematic diagram of prototype CAM.

and unattached background radon/thoron progeny. After passing through the condenser, the aerosol flows into the gap between the filter and detector. In the current configuration, the detector can be as large as 49 mm diameter and the diameter of the open area of the filter can be as large as 42 mm. The filter is mounted in a special holder which, in turn, is inserted into the sampler in a drawer. During sampling, the filter is sealed in place with a mechanical cam arrangement. The flow through the CAM is controlled with a critical flow venturi (Wright, 1954) and monitored with a mass flow meter (Sierra Instruments, Inc., Carmel Valley, CA).

III. Methodology and Test Apparatus

The wind tunnel used in the testing, Figure 2, has a basic cross section of 610 mm × 610 mm which is expanded to 1000 mm × 1000 mm at the test section. The expansion is designed to reduce blockage effects in the test section. Aerosol was generated with a vibrating jet atomizer (Berglund and Liu, 1973) from a mixture of nonvolatile oleic acid in ethanol. A fluorescent analytical tracer, sodium fluorescein, was added to the oleic acid in a ratio of 10% (m/V). During testing, samples of the oleic acid aerosol were collected on oilphobic glass slides and examined under a light microscope to determine the size. The resulting observed sizes were converted to AED by using the flattening factor of Olan-Figueroa et al. (1982) and the calculated density of the oleic acid/sodium fluorescein mixture.

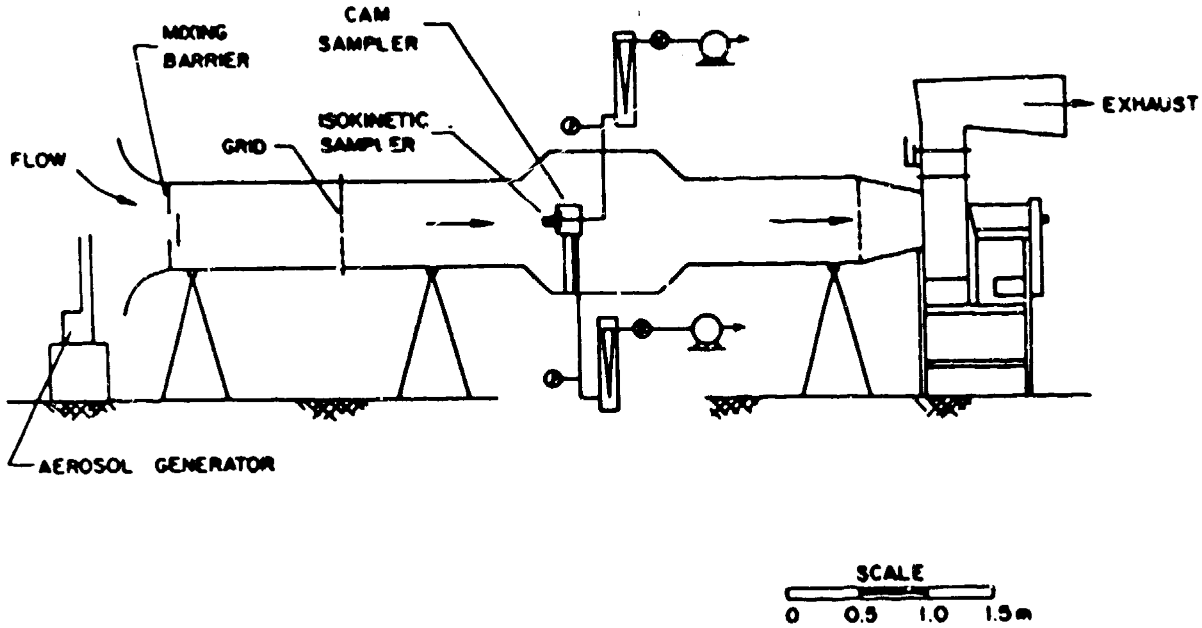


Figure 2. Wind tunnel used to characterize aerosol sampling attributes of the prototype CAM sampler.

Freshly formed aerosol was introduced into the wind tunnel through a mixing barrier and a grid plate. The purpose of these elements is to obtain a uniform aerosol concentration profile over the center 2/3 of the wind tunnel. In the test section, the aerosol was simultaneously sampled with the CAM prototype and an isokinetic probe fitted with a filter collector. At the completion of a test, the filters were removed from the CAM and isokinetic probe and brought to an analysis laboratory where the sodium fluorescein was eluted and subsequently quantified. Aerosol penetration, P, through the CAM was calculated from:

$$\phi = \frac{m_{f,c}\, Q_{iso}}{m_{f,iso}\, Q_c} \quad (1)$$

where: $m_{f,c}$ and $m_{f,iso}$ are the masses of fluorescein collected by the CAM and isokinetic filters, respectively; and, Q_c and Q_{iso} are the flow rates through the two samplers. At least triplicate tests were run at each condition in order to provide a measure of the reproducibility of the experiments.

The tests which involved determination of filter uniformity consisted of operating the CAM sampler in the wind tunnel for a period of time sufficient to collect an easily analyzable quantity of fluorescein, cutting the filter into 20 subsamples and then quantifying the fluorecein on each of the subsamples. Triplicate experiments were conducted at each test condition.

The concept of using fractionators to remove high mobility background alpha-emitters from the aerosol size distribution was tested with the apparatus shown in Figure 3. High grade uranium ore was placed in a 200 L vessel as a means of generating radon daughters. Filtered room air was admitted into the vessel and then drawn into two commercially available CAM samplers. One of the CAM samplers had no fractionator and the other was fitted with an inlet which contained a screen and/or an electrostatic precipitator. The resulting energy spectra were analyzed for radon daughters in selected regions of interest. We also conducted experiments with partially aged radon daughters, where the radon daughters were given the opportunity to attach themselves to aerosol particles in room air. For these latter tests, the air exposed to the high grade uranium ore was discharged directly into a (200 m^3) laboratory environment. The two CAM samplers were positioned about 5 m from the radon daughter source. Again, comparisons were made between the counts accumulated with the fractionating CAM those detected with the unmodified CAM.

IV. Results

Since one of the key parameters in the design of a CAM sampler is the spacing between filter and detector, a set of tests was conducted to determine its effect of spacing upon aerosol penetration. Here, the sampler was operated at a flow rate of 57

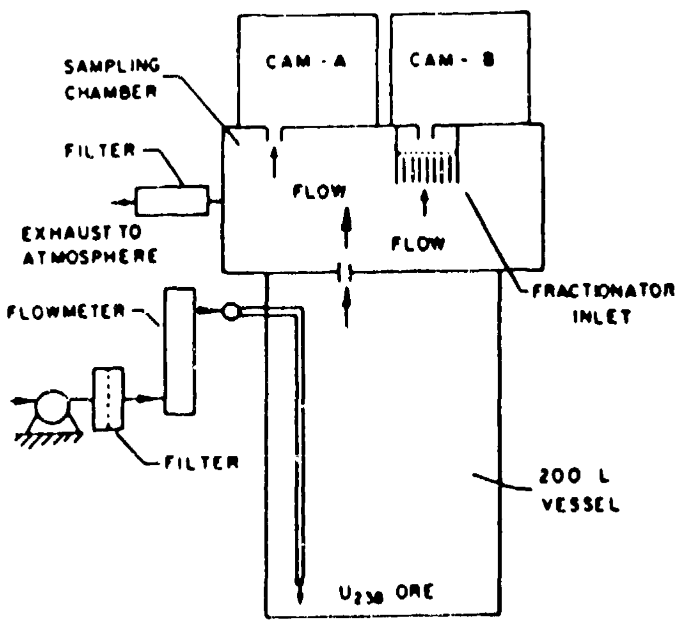


Figure 3. Test apparatus used to characterize the transmission of radon daughters through a fractionating inlet. CAM A had no special inlet and was used to provide a comparative reference with the fractionating inlet on CAM B.

L/min (2 cfm) in a wind speed of 1 m/s and challenged with 10 μm AED aerosol particles. The gap between filter and detector was set at various levels from 3.3 to 7.1 mm. It should be noted, however, that the actual minimum gap through which the aerosol flows is 2.03 mm smaller than the filter/detector gap due to the presence of the filter holder and the detector clamping ring. The results of these experiments, Table 1, show the penetration to be unaffected by filter/detector gap over the range of values tested. For all experiments we noted the penetration of 10 μm AED particles was between 85 and 87%. As a conservative step, we selected a gap of 4.6 mm for use in the CAM design.

The effect of wind speed on sampler performance is given in Table 2. The flow rate was 57 L/min, the particle size was 10 μm AED, and the wind speed was set at 0.3, 1, and 2 m/s. The values of penetration corresponding to these three wind speeds are 83, 87 and 88% with a standard deviation of about 3%, which indicates there is no substantial speed dependency over the range of speeds tested. No screen-type inlet fractionator was used during these tests.

Table 1. Penetration of aerosol through the prototype CAM sampler as a function of gap between filter and detector. Wind speed = 1.0 m/s, particle diameter = 10 μm AED, flow rate = 57 L/ min. No inlet screen. Minimum gap between filter holder and detector holder is 2.03 mm less than the filter/detector gap.

Filter/ Detector Gap (mm)	Penetration (percent)	Std. Dev. (percent)
3.3	87.1	0.9
3.9	85.9	1.7
4.6	87.1	2.7
5.8	86.3	1.7
7.1	86.7	1.7

Table 2. Penetration of aerosol as a function of wind speed. Flow rate = 57 L/ min, particle diameter = 10 μm AED. No inlet screen.

Wind Speed, m/s	Penetration (percent)	Std. Dev. (percent)
0.3	83.2	4.0
1.0	87.1	2.7
2.0	88.1	3.1

The variation of aerosol penetration with flow rate at a wind speed of 1 m/s is shown in Figure 4. Essentially 100% of 5.4 μm AED aerosol particles penetrate from the free stream to the sampling filter. At a size of 10 μm AED, the penetration values are 92%, 87% and 79% corresponding to flow rates of 28, 57, and 85 L/min, respectively. The cutpoint particle size (AED for which the penetration is 50%) is 17 μm for a flow rate of 57 L/min. No inlet screen was used during these tests.

The effect of including a fine mesh inlet screen with 0.11 mm diameter wires is shown in Figure 5. For those tests, the CAM was operated at a flow rate of 57 L/min in a wind speed of 1 m/s. It may be noted that the presence of the screen reduces the penetration of 10 μm AED particles from 87 to 71%. The cutpoint particle size is approximately 13 μm AED when the screen is used. Research is continuing on the design and placement of the screen elements.

Filter uniformity data for 10 μm AED aerosol particles are summarized in Table 3. The sampler was operated a flow rate of 57 L/min in a wind speed of 0.3 m/s for these tests. The locations on a filter from which each of the 20 subsamples were cut are identified in the drawing shown in Table 3. Test results have been normalized to a mean areal deposition of unity for each test. The coefficient of variation of areal deposition values is 7% for tests conducted either with or without a fine mesh screen over the inlet. Other tests which were conducted at flow rates of 28 and 85 L/min showed coefficients of variation of 4% and 8%, respectively.

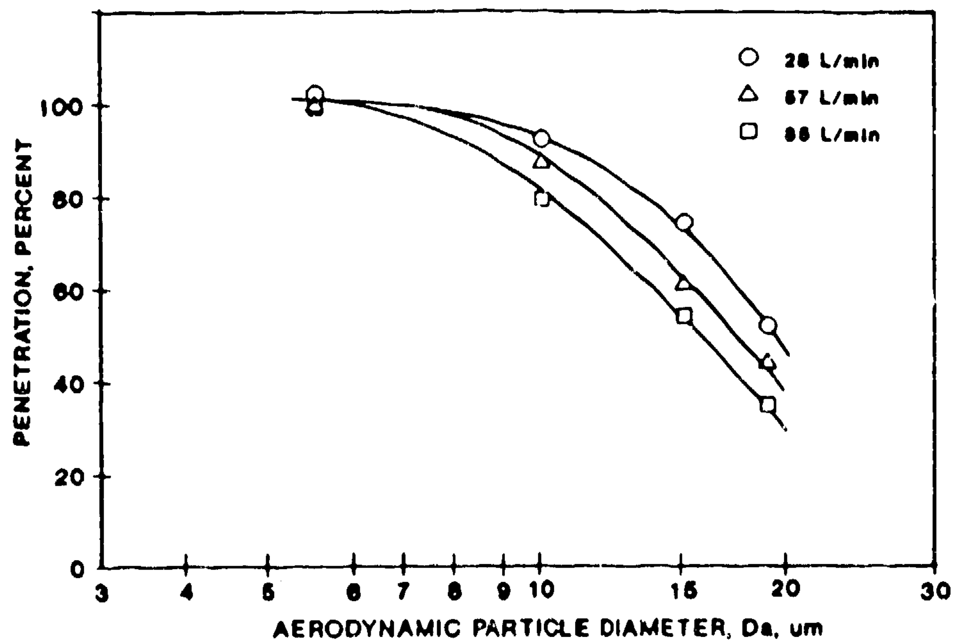


Figure 4. Effect of flow rate upon aerosol penetration. Wind speed = 1 m/s.

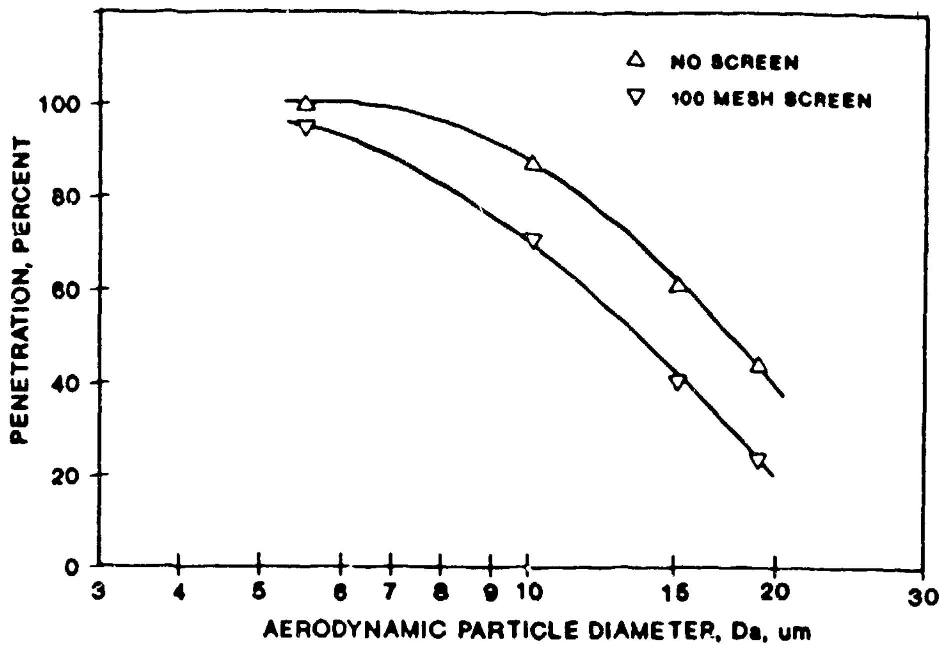


Figure 5. Effect of a 100 mesh inlet screen upon aerosol penetration. Flow rate = 57 L/min, Wind speed = 1 m/s.

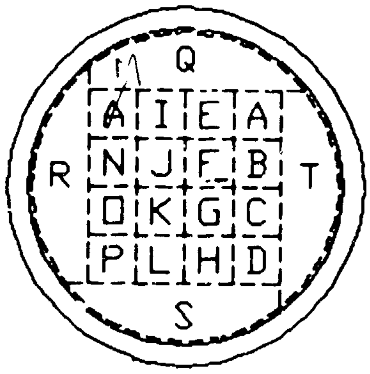
Tests conducted to characterize the removal of freshly formed radon daughters showed the combination of a fine mesh screen and an electrostatic condenser eliminated 95% of the background radionuclides in the region of interest. Correspondingly, the removal of partially aged radon daughters was 33%.

V. Discussion

The CAM sampler prototype which is reported herein has a cutpoint which is greater than 10 μm AED when operated at a flow rate of 57 L/min with or without an inlet screen. We believe that a cutpoint of at least 10 μm should be used as a performance criteria for CAM samplers since this size can penetrate to the thoracic region of the human lung tree (ACGIH, 1985) and since particles of this size, or larger, can easily be generated under certain accident or release scenarios (Elder et al., 1974; Perrin, 1987; Ballinger et al., 1988). From the standpoint of alarm considerations, it is also important that larger particles be effectively collected. The current standard for Pu-239 corresponds to the amount of alpha radiation which is emitted by a single particle of approximately 10 μm AED over an 8-hour period. Inadequate collection of larger particles could affect the ability of an instrument to correctly signal an alarm.

The performance of the prototype CAM in the collection of 10 μm AED particles at a flow rate of 57 L/min is relatively unaffected by

Table 3. Filter uniformity areal deposition. Values are normalized to a mean of unity. Wind speed = 0.3 m/s, particle diameter = 10 μm AED and flow rate = 57 L/min. Three replicates at each condition. The ± values are standard deviations.



Sub-sample	Without an Inlet Screen		With a Fine Mesh Screen On Inlet	
A	0.96	±0.05	0.94	±0.03
B	0.93	±0.01	0.88	±0.06
C	1.00	±0.03	0.89	±0.05
D	1.04	±0.08	1.00	±0.05
E	0.95	±0.05	0.96	±0.02
F	0.94	±0.03	1.02	±0.02
G	1.01	±0.01	1.04	±0.03
H	1.06	±0.07	1.05	±0.05
I	0.95	±0.01	1.00	±0.02
J	0.93	±0.02	1.10	±0.05
K	1.03	±0.02	1.08	±0.06
L	1.02	±0.04	1.08	±0.02
M	1.13	±0.03	0.97	±0.02
N	1.09	±0.08	1.08	±0.06
O	1.12	±0.02	1.04	±0.03
P	1.07	±0.05	1.04	±0.02
Q	0.90	±0.03	0.94	±0.04
R	0.94	±0.02	0.98	±0.03
S	1.01	±0.03	1.03	±0.03
T	0.91	±0.02	0.89	±0.03
Coefficient of Variation, %	0.07		0.07	

wind speed over the range of 0.3 to 2 m/s, showing penetration values of approximately 85% for these conditions. In clean room laboratory environments, the recommended mean air speed is approximately 0.5 m/s (ASHRAE, 1987), a value which is encompassed by the range of test conditions. It is anticipated that sampling biases would be primarily associated with high wind speeds (Durham and Lundgren, 1980) so it is expected the prototype CAM should perform well as an area sampler in a laboratory where the air velocity is within the tested range. Also, flow rate over the range of values of 28 to 85 L/min does not have a substantial effect on the sampling performance; thus, if a sampler is operated at a flow rate which deviates from the design condition of 57 L/min, the characteristics of the sample will not be greatly affected.

The uniformity of deposits of 10 μm AED aerosol particles on sampling filters is 7%, as represented by the coefficient of variation of 20 subsamples cut from each test filter. This is a lower values than noted in earlier tests with two commercially available CAM samplers which were able to transmit significant fractions of 10 μm AED aerosol particles (McFarland et al., 1990).

Preliminary experiments with a prototype inlet which fractionates high mobility background alpha-emitters show 95% of freshly formed radon daughters can be removed from the size distribution prior to collection of the aerosol by the sampling filter. When the radon daughters were allowed to become partially attached to ambient aerosol particles, the removal dropped to 33%. For certain sampling situations, the use of inlet fractionators could provide a relative enrichment of the TRU fraction of the aerosol and thereby improve the quality of alarm signals.

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